Investigation of the Photocatalytic Degradation of Ethanol and Acetone

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Abstract: In-situ transmission Fourier-transform infrared spectroscopy has been used to study the photocatalytic oxidation of acetone, ethanol and the interaction between acetone and ethanol. Compared with the degradation of acetone alone, it cannot be described by Langmuir-Hinshelwood equation in presence of ethanol. The presence of ethanol reduces the initial degradation rate of acetone and the inhibition increases with increasing of ethanol in the system. Acetone also inhibits the degradation of ethanol but it still can be described by the L-H equation. Acetaldehyde in the system comes from the degradation of ethanol, the behavior of production and consumption of which is affected by the amount of ethanol and acetone in the mixture. Temperature significantly affects the degradation of organic compounds in the mixture. Increasing the temperature accelerates the degradation of ethanol and acetone as well as the degradation of acetaldehyde, an intermediate produced in the system. The flux of the reaction system has little effect on the photocatalytic process of organic matter.

Key words: photocatalytic degradation; ethanol; acetone

1. INTRODUCTION

People generally spend 88% of their time in indoor environment on average [1]. The quality of indoor air has great effect on human health in terms of lengthened exposure to pollutants by inhalation. Volatile organic compounds (VOCs) are familiar indoor pollutants. Photocatalytic degradation of VOCs is promising way to lighten air pollution and much work has been done using single organic compound [1-8], while the studies on the interaction between organic compounds during photocatalytic decomposition are limited. In this paper, the photocatalytic oxidation of acetone, ethanol and the interaction between them during photocatalysis process have been investigated using a Fourier-transform infrared spectroscopy. Ethanol and acetone are chosen as target pollutants because ethanol is a well-known organic compound and acetone is a popular indoor air pollutant.

2. EXPERIMENTAL

2.1 Reactor and gas generator

Fig.1 shows the schematic diagram of the experimental set-up for the study. It consists of a reactor and an air stream circulation system. The reactor includes a 20-W UV lamp with a 254nm wavelength illumination and a glass tube with a 66mm inside diameter and 620mm long coated with TiO_2 powder in advance. They are located concentrically with 5mm radial clearance. The ports on the left are used to put in the air containing reactants before each experimental run, which is produced by the assembly shown in Fig.2. During experiment the valve No.1 and No.3 are closed and the valve No.2 is open. Sampling ports are on the right. They are connected with a Fourier transform–infrared radiation (FTIR) spectroscopy apparatus which is used to monitor the reactants and products. The valve No.5 is closed while No. 4 and No. 6 are open for air to go into FTIR monitoring system during experiment. The fan in the system is used to control the flux of the air.

Before experiment the set-up is washed for 10min by fresh air at the rate of 500ml/min and then the air with pollutants is pressed in the set-up. After valves No.1 and No. 3 are turned off the No.2 is

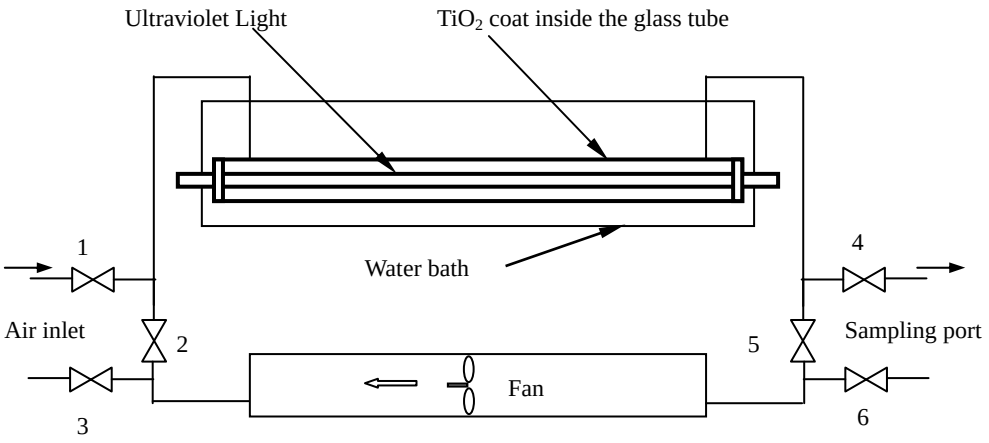

Fig.1 Schematic diagram of the experimental set-up

turned on. The fan in the system starts to work and No. 4 and No. 6 on the sample ports are open. After the system is stable the UV-lamp is turned on and the photocatalytic degradation of organic compounds is enabled. The data is recorded by a FTIR (made in the PERKIN-ELME Company) per 10 min, which has been quantified in advance by a gas chromatographic analysis (SP2100, made in Beijing). Temperature in the reactor is regulated through the water temperature in the water bath. The flux of air in the system is adjusted through the fan.

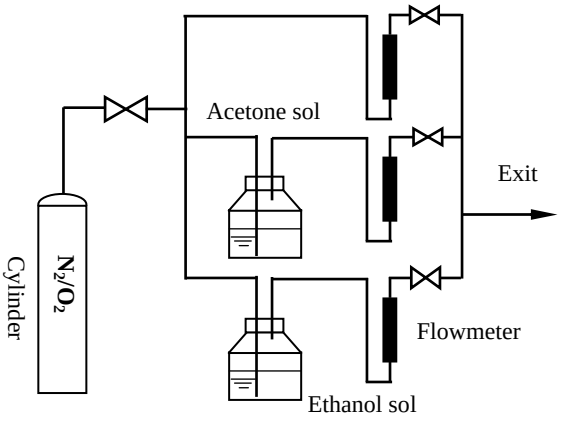

Fig.2 Generator of the air with ethanol and acetone

Fig.2 presents the system to generate the air containing the ethanol, acetone or the mixture of them. Mock air in the cylinder consists of N_2 and O_2. It is pressed to experimental set-up in Fig.1 by three ways. Two of the ways are used to produce the air containing pollutants and one is used for cleaning the set-up before experiment. The bottles in the ways are filled with ethanol and acetone solutions respectively. Compressed air through them can entrain ethanol and acetone molecules and thus the air containing pollutants is produced.

2.2 Reagents and catalyst preparation

Ethanol and acetone used as reagents is produced from the system shown in Fig.2. The mass percentage of the solution of ethanol is 99.7% and that of acetone is 99.5%.

The catalyst used in the study is a TiO_2 powder (Degussa P-25). It is a mischcrystal powder and the mass ratio of anatase to rutile is 4:1. The average particle diameter is 21 nm and the surface area is $50m^2/g$. A water suspension with the TiO_2 powder, the mass percentage of which is 2.5% is made in advance. During the suspension making process a magnetic stirring apparatus has been used to stir the suspension for 30min before it is used to form the coat on a glass tube. Filled with the water suspension, the glass tube is loaded with TiO_2 on its side by soaked and wiggled repeatedly and then dried at 373K for 1 h in a drying oven heated by electricity. It is soaked and dried again and again until 2.05g TiO_2 is coated, i.e., the density of TiO_2 loaded on the tube is $3.24 \times 10^{-3} g/cm^2$. The amount of TiO_2 loaded is determined by the weight difference before and after the coating procedure. By the way, when the density of TiO_2 on the side of the tube is over $3.2 \times 10^{-3} g/cm^2$ no light could permeate it [6].

3. RESULTS AND DISCUSSION

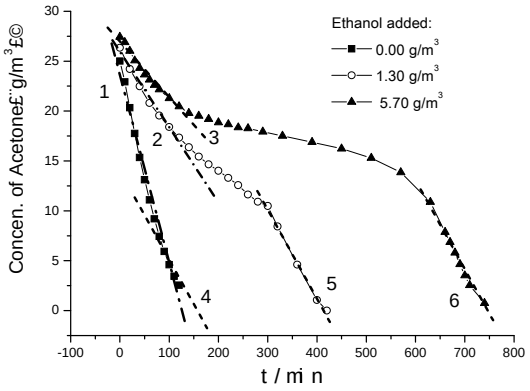

Fig. 3 Degradation of acetone in the presence and the absence of ethanol

3.1 Degradation of acetone in the presence of ethanol

Fig.3 demonstrates the degradation of acetone in the presence or absence of ethanol. The experiments were carried out at the average temperature of 14℃ and the air flux of 500ml/min. The slope of the short dash lines drawn in Fig.3 denotes the mean reaction rate of acetone, the values of which are given in Table 1.

Tab. 1 Mean slope rate of the lines in Fig. 3

No.	1	2	3	4	5	6
Values	0.210	0.080	0.064	0.100	0.100	0.100
Error	±0.005					

It is clear that during the degradation of single acetone the reaction rate can be described approximately by the rate of slope of dash line 1, while it can characterized by the slope coefficient of line 4 when the concentration of acetone in the system is less than 8g/m^3. The rate of slope of line 1 is the steepest among lines 1, 2 and 3 indicating that the initial degradation rate of acetone in single case is the fastest. The presence of ethanol reduces the initial degradation rate of acetone and the inhibition of ethanol increases with increasing the amount of ethanol in the system.

With the development of the photo-catalytic degradation, acetone concentration decreases against time gradually. Before it reaches 8g/m^3 the inhibitory action of ethanol has existed. It can be seen in Fig. 3 that the higher the concentration of ethanol the lower the degradation rate of acetone. After the concentration of acetone falls to 8g/m^3, the degradation rate can be expressed by the slope coefficient of parallel Line 4, 5 and 6, which is less that of Line 1. This result indicates the effect of ethanol disappear hereafter. In a word, compared with the degradation of acetone alone, it cannot be described by Langmuir-Hinshelwood equation in presence of ethanol. The presence of ethanol reduces the initial degradation rate of acetone and the inhibition is increased with the increase of ethanol in the system.

3.2 Degradation of ethanol in the presence of acetone

Fig.4 presents the concentration of the ethanol against time with and without acetone. The average initial concentration of ethanol is 6.14 g/m^3. The initial concentration of acetone is 0, 12.54 g/m^3 and 27.42g/m^3 respectively, the temperature is 4℃, and the flow rate is 500ml/min.

As can be seen In Fig. 4, the concentration of the ethanol decreases gradually against time. The presence of acetone inhibits the degradation of ethanol. The higher the initial concentration of acetone is, the stronger the inhibition is. Nevertheless, the degradation of ethanol still can be described by L-H equation. The existence of acetone makes the time for degradation of ethanol longer.

3.3 Production of acetaldehyde

Fig.5 shows the acetaldehyde concentration produced during the degradation of ethanol, acetone and the mixture of them. It can be observed that only

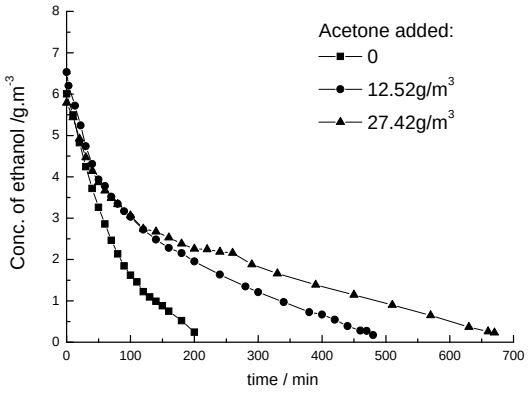

Fig.4 Ethanol concentration against time in the presence and the absence of acetone

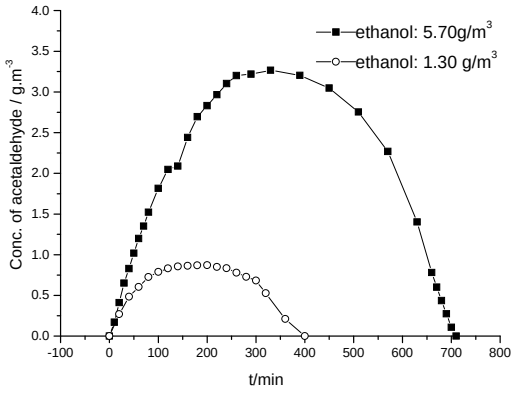

Fig.5 Acetaldehyde produced during the degradation of ethanol in presence of acetone of 26g/m^3

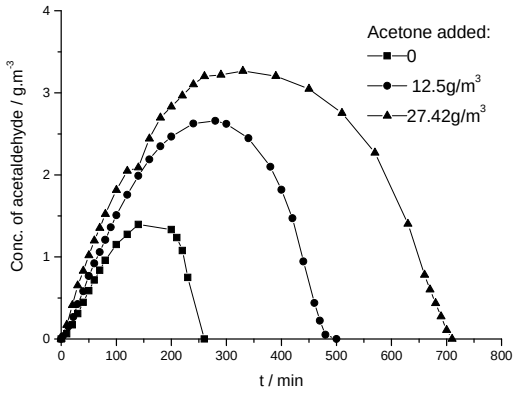

Fig.6 Effect of the acetone in presence of ethanol of 6g/m^3

two acetaldehyde curves appear. There is no acetaldehyde produced during the photo-catalytic degradation of acetone. It is concluded that the acetaldehyde comes from the degradation of ethanol. The behavior of production and consumption of acetaldehyde is affected by the amount of ethanol in the mixture. With the increase of the initial concentration of ethanol the peak concentration of acetaldehyde increases and the time for acetaldehyde to be exhausted is prolonged.

Acetone affects the production and consumption of acetaldehyde (See Fig. 6). The amount of acetone in the system also affects the behavior of production and consumption of acetaldehyde. Increasing the initial concentration of acetone in the system can raise the peak concentration of acetaldehyde and extend the subsistence time of acetaldehyde. It can be understood since the acetaldehyde concentration in the reaction system results from the balance of the two reactions: one is the reaction that ethanol is oxidized to acetaldehyde and the other one is that acetaldehyde is photo-catalytically degraded. The results above indicate that the presence of acetone inhibits the degradation of acetaldehyde, resulting that the increase of peak concentration acetaldehyde and the prolongation of the subsistence time.

3.4 Effect of temperature

Fig.7, 8 and 9 illustrate the ethanol, acetone and acetaldehyde concentrations against time at the temperature of 14℃, 31℃ and 43℃ respectively. The flow rate is 500ml/min. In Fig.7 it can be seen

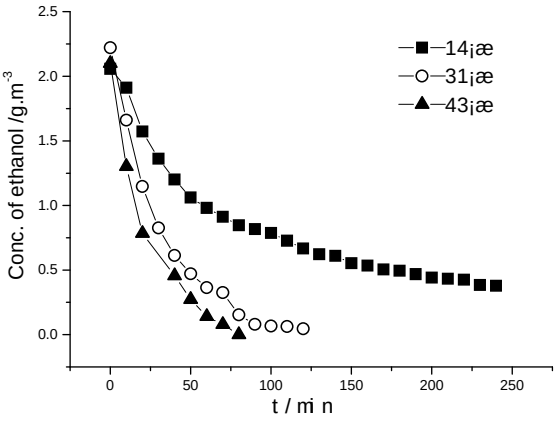

Fig.7 Effect of temperature on the ethanol degradation

that the higher the temperature is, more quickly the ethanol concentration decreases.

In Fig.8, the degradation rate of acetone in the system is also found to increase with increasing temperature. With the increasing of temperature the peak concentration and the time for acetaldehyde to be exhausted are reduced (See Fig. 9). The results indicate that temperature affects greatly the degradation of organic compounds. Raising the temperature of the reaction system can accelerate the degradation of ethanol, acetone as well as the degradation of acetaldehyde, an intermediates produced in the system.

3.5 Effect of flux

Fig.10, 11 and 12 present the concentration of ethanol, acetone and acetaldehyde against time in the

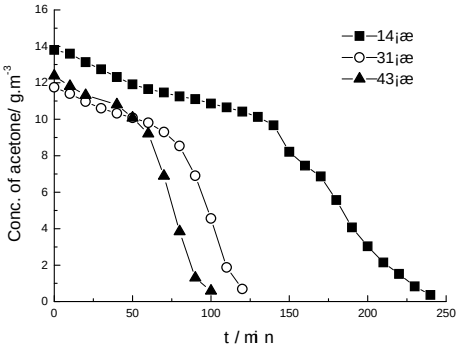

Fig.8 The effect of temperature on acetone degradation

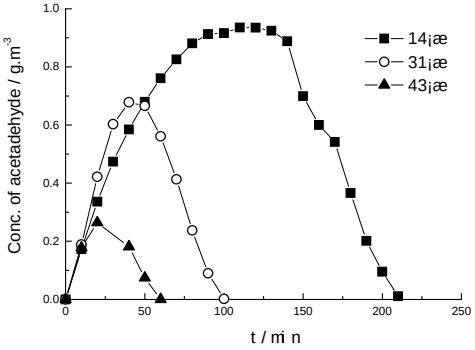

Fig.9 The effect of temperature on acetaldehyde produced.

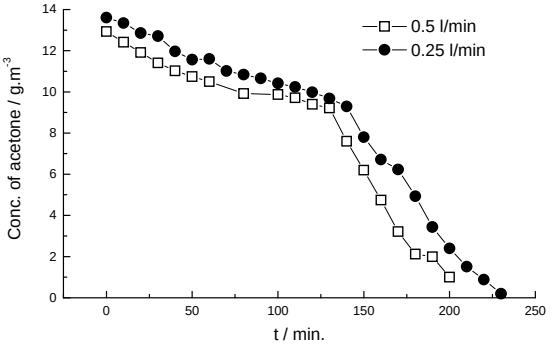

Fig.10 Effect of flow rate on the degradation of acetone

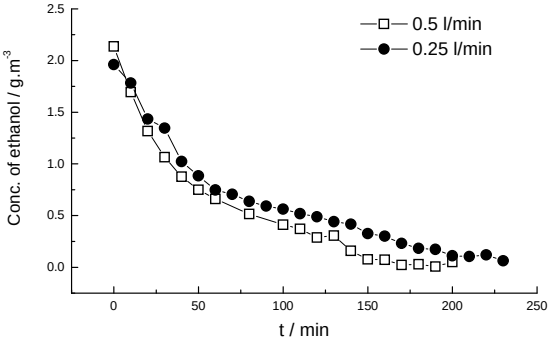

Fig.11 Effect of flow rate on the degradation of ethanol

reaction system at the flow rate of 0.5 and 0.25l/min. Temperature is 14℃. As can be seen, the curves between the two flow rates are overlapped and trends almost in the same way. The results indicate that the degradation rates of ethanol, acetone and acetaldehyde at the different flow rates are proximal or the flux in the reaction system has little effect on the photo-catalytic degradation of organic matter. It can be easily understood that although increasing the flux in the batch reactor means the increase of the circling times of the gas, it decreases the contact time of gas with TiO_2 film at every circling. Therefore flux or the flow rate of the system has little effect on the photo-catalytic degradation of organic matter.

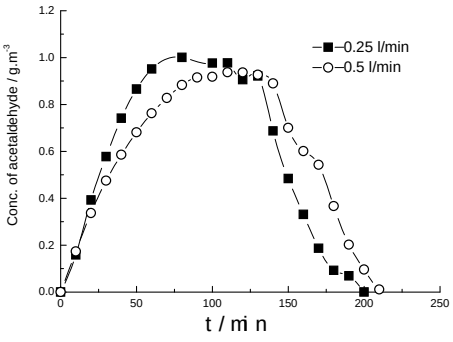

Fig.12 Effect of flow rate on the degradation of acetaldehyde

4. CONCLUSIONS

The photocatalytic oxidation of acetone, ethanol and the interaction between them during photocatalysis process are investigated using a Fourier-transform Infrared Spectroscopy. Compared with the degradation of acetone alone, it cannot be described by L-H equation in presence of ethanol. The presence of ethanol reduces the initial degradation rate of acetone and the inhibition increases with increasing the amount of ethanol in the system. Acetone also inhibits the degradation of

ethanol while it still can be described by L-H equation. Acetaldehyde in the system comes from the degradation of ethanol, the behavior of production and consumption of which is affected by the amount of ethanol and acetone in the mixture. Temperature affects greatly the degradation of organic compounds. Raising temperature accelerates the degradation of ethanol, acetone as well as the degradation of acetaldehyde, an intermediate produced in the system. The flux of the reaction system has little effect on the photo-catalysis process of organic matter.

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